

FORM PTO-1390 (Modified) (REV 11-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER 205744US0PCT
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 09/830149
INTERNATIONAL APPLICATION NO. PCT/EP99/07899	INTERNATIONAL FILING DATE 19 OCTOBER 1999	PRIORITY DATE CLAIMED 21 OCTOBER 1998		
TITLE OF INVENTION PARAFFIN DISPERSANTS HAVING A LUBRICITY EFFECT FOR MINERAL OIL MIDDLE DISTILLATES				
APPLICANT(S) FOR DO/EO/US Bernd WENDEROTH, et al.				
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:				
<ol style="list-style-type: none"> 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (24) indicated below. 4. <input checked="" type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31). 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371 (c) (2)) <ol style="list-style-type: none"> a. <input type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau). b. <input checked="" type="checkbox"/> has been communicated by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). <ol style="list-style-type: none"> a. <input checked="" type="checkbox"/> is attached hereto. b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4). 7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) <ol style="list-style-type: none"> a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> have been communicated by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input checked="" type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). 10. <input checked="" type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)). 11. <input type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409). 12. <input checked="" type="checkbox"/> A copy of the International Search Report (PCT/ISA/210). 				
Items 13 to 20 below concern document(s) or information included:				
<ol style="list-style-type: none"> 13. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 14. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 15. <input checked="" type="checkbox"/> A FIRST preliminary amendment. 16. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. 17. <input type="checkbox"/> A substitute specification. 18. <input type="checkbox"/> A change of power of attorney and/or address letter. 19. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825. 20. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4). 21. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4). 22. <input type="checkbox"/> Certificate of Mailing by Express Mail 23. <input checked="" type="checkbox"/> Other items or information: 				
Request for Consideration of Documents in International Search Report PCT/IB/304 / Notice of Priority/ PCT/IB/308 / Amended Sheets (pages 14, 15 and 15a)				

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR
09/830149INTERNATIONAL APPLICATION NO.
PCT/EP99/07899ATTORNEY'S DOCKET NUMBER
205744US0PCT

24. The following fees are submitted.:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :

- | | |
|--|-----------|
| <input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO | \$1000.00 |
| <input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO | \$860.00 |
| <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO | \$710.00 |
| <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) | \$690.00 |
| <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) | \$100.00 |

CALCULATIONS PTO USE ONLY**ENTER APPROPRIATE BASIC FEE AMOUNT =**

\$860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)).

 20 30

\$0.00

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	9 - 20 =	0	x \$18.00	\$0.00
Independent claims	1 - 3 =	0	x \$80.00	\$0.00
Multiple Dependent Claims (check if applicable).				\$0.00

TOTAL OF ABOVE CALCULATIONS = \$860.00

- Applicant claims small entity status. (See 37 CFR 1.27). The fees indicated above are reduced by 1/2.

\$0.00

SUBTOTAL = \$860.00

Processing fee of \$130.00 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f)).

 20 30

\$0.00

TOTAL NATIONAL FEE = \$860.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable).

 \$0.00**TOTAL FEES ENCLOSED =** \$860.00

Amount to be: \$

refunded \$

charged \$

- a. A check in the amount of \$860.00 to cover the above fees is enclosed.
- b. Please charge my Deposit Account No. _____ in the amount of _____ to cover the above fees. A duplicate copy of this sheet is enclosed.
- c. The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 15-0030. A duplicate copy of this sheet is enclosed.
- d. Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:



22850

(703) 413-3000

Surinder Sachar
Registration No. 34,423

SIGNATURE

Norman F. Oblon

NAME

24,618

REGISTRATION NUMBER

Apr. 11 2001

DATE

09/850147

JC03 RCP/TM PCT/PTO

23 APR 2001

205744US0PCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF: :

BERND WENDEROTH ET AL : ATTN: APPLICATION DIVISION

SERIAL NO: NEW US PCT APPLN :
(BASED ON PCT/EP99/07899)

FILED: HEREWITH :

FOR: PARAFFIN DISPERSANTS HAVING
A LUBRICITY EFFECT FOR
MINERAL OIL MIDDLE DISTILLATES

PRELIMINARY AMENDMENT

ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

SIR:

Prior to examination on the merits, please amend the above-identified application as follows.

IN THE CLAIMS

Please cancel Claims 3-11 and add the following claims:

12. (New) A mixture as claimed in claim 1, wherein the reaction product (a) is an amide, amidoammonium salt or ammonium salt in which no carboxyl group, one carboxyl group or a plurality of carboxyl groups is or are converted into amido groups.

13. (New) A mixture as claimed in claim 1, wherein the secondary amine of the component (a) is of the formula HNR₂, where R is straight-chain or branched C₁₀₋₃₀-alkyl.

14. (New) A mixture as claimed in claim 1, wherein the primary alkylamine of the component (b) is a C₈₋₃₀-alkylamine.
15. (New) A mineral oil middle distillate containing a mixture as claimed in claim 1.
16. (New) A mineral oil middle distillate as claimed in claim 15, further comprising flow improvers, conductivity improvers, corrosion-inhibiting additives, lubricity additives, antioxidants, metal deactivators, antifoams, demulsifiers, detergents, cetane number improvers, dyes, fragrances or mixtures thereof.
17. (New) A concentrate containing 10-60% by weight, based on the total amount of the concentrate, of a mixture as claimed in claim 1, dissolved in the hydrocarbon solvent.
18. (New) A mineral oil middle distillate as claimed in claim 15, further comprising flow improvers based on ethylene/vinyl carboxylate copolymers.

REMARKS

Claims 1, 2 and 12-20 are active in the present application. Support for Claims 12-20 is found in Claims 3-11 and the specification. No new matter is added. An action on the merits and allowance of the claims is solicited.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,
MAIER & NEUSTADT, P.C.



Norman F. Oblon
Attorney of Record
Registration No. 24,618

Daniel J. Pereira, Ph.D.
Registration No. 45,518



22850

(703) 413-3000
Fax #: (703)413-2220
NFO/DJPER/js

I:\user\DJPER\205744US-PR.wpd

Marked-Up Copy
Serial No: _____
Amendment Filed on: _____

IN THE CLAIMS

Claims 3-11 (Canceled.

12. (New)

13. (New)

14. (New)

15. (New)

16. (New)

17. (New)

18. (New)

"as originally filed"

**Paraffin dispersants having a lubricity effect
for mineral oil middle distillates**

5 The present invention relates to mixtures suitable as paraffin dispersants having a lubricity effect, their use in mineral oil middle distillates, mineral oil middle distillates of this type and concentrates for this purpose.

10 Mineral oil distillates, in particular middle distillates, such as gas oils, diesel oils or light fuel oils, which are obtained by distillation from mineral oils, have different paraffin contents,

15 depending on the origin of the crude oil. At relatively low temperatures, solid paraffins separate out (cloud point, CP). On further cooling, the lamellar n-paraffin crystals form a type of "house of cards structure" and the middle distillate sets although the predominant

20 part of the middle distillate is still liquid. The flowability of the middle distillate fuels is considerably impaired by the precipitated n-paraffins in the temperature range between cloud point and pour point; the paraffins block filters and result in a

25 nonuniform supply of fuel to the combustion units or completely stop said supply. Similar faults occur in the case of light fuel oils.

30 It has long been known that the crystal growth of the n-paraffins in the combustion or power fuels obtained from mineral oil distillates can be modified by suitable additives. Effective additives prevent middle distillates from becoming solid at temperatures of only a few degrees Celsius below the temperature at which

the first paraffin crystals separate out. Instead, fine, well crystallized, separate paraffin crystals form, which crystals pass through filters in motor vehicles and heating systems or at least form a filter
5 cake which is permeable for the liquid part of the middle distillates, so that trouble-free operation is ensured. According to European standard EN 116, the efficiency of the flow improvers is expressed indirectly by measuring the cold filter plugging point
10 (CFPP).

Ethylene/vinyl carboxylate copolymers have long been used as flow improvers. The disadvantage of these additives is that, owing to their higher density
15 compared with the liquid part, the precipitated paraffin crystals tend to settle out to an increasing extent on the bottom of the container on storage. Consequently, a homogeneous low-paraffin phase forms in the upper part of the container and a two-phase
20 paraffin-rich layer at the bottom. Since the middle distillate is generally taken off slightly above the bottom of the tank, both in the vehicle tanks and in storage or delivery tanks of the mineral oil dealers, there is a danger that the high concentration of solid
25 paraffins will block filters and metering means. This danger is all the greater the greater the amount by which the storage temperature is below the precipitation temperature of the paraffins, since the amount of precipitated paraffin increases with
30 decreasing temperature.

These problems can be reduced by the additional use of paraffin dispersants (wax antisettling additives).
35 Thus, EP-A-0 398 101 describes reaction products of aminoalkylene polycarboxylic acids with long-chain, secondary amines as paraffin dispersants for mineral oil middle distillates. However, the effect is not sufficient in all mineral oil middle distillate

compositions, particularly when they have a low sulfur content of less than 500 ppm.

DE-A-11 49 843 describes the use of maleamic acids and 5 their amine salts obtained from primary amines and maleic anhydride as corrosion inhibitors and stability improvers for mineral oil distillates as well as for preventing sediment formation. EP-A-0 106 234 describes the use of amine salts of maleamic acids obtained from 10 primary amines and maleic anhydride as corrosion inhibitors for the storage and the transport of crude oils.

Since October 1996, only low-sulfur middle distillates 15 having a sulfur content of not more than 500 ppm may be used as diesel fuels in the European Union, which fuels, owing to said sulfur content, help to achieve exhaust gases having a lower pollutant content. From the year 2000 onward, the maximum permissible sulfur 20 content of diesel fuels in the European Union may not exceed 350 ppm. However, such diesel fuels have a substantially reduced lubricity, which can in some cases lead to high mechanical wear in distributor injection pumps of diesel engines.

25 The lubricity of low-sulfur diesel fuels can be improved by adding lubricity additives. According to standard CEC F-06-A-96, the efficiency is determined by the HFRR test (High Frequency Reciprocating Rig test) 30 by determining the wear size WS1.4 in μm at 60°C; the smaller the WS1.4, the lower is the wear and the better is the lubricity.

There are numerous patent applications for products 35 which can improve the lubricity of low-sulfur diesel fuels. WO 95/33805 states that flow improvers and paraffin dispersants, including reaction products according to EP-A-0 398 101, can improve the lubricity of middle distillates. However, the lubricity effect is

not sufficient in many mineral oil middle distillate compositions.

It is an object of the present invention to provide
5 improved products which ensure improved flowability of
mineral oil middle distillates at low temperature by
virtue of the fact that they have a dispersing effect
such that settling of precipitated paraffins is delayed
or prevented, and which simultaneously contribute to
10 improved lubricity of the mineral oil middle
distillates.

We have found that this object is achieved, according
to the invention, by a mixture containing
15

- (a) from 5 to 95% by weight of at least one reaction product of a poly(C_{2-20} -carboxylic acid) having at least one tertiary amino group with secondary amines and
20
- (b) 5-95% by weight of at least one reaction product of maleic anhydride and a primary alkylamine.

25 The present invention also relates to the use of these mixtures as additives for mineral oil middle distillates, in particular as paraffin dispersants and lubricity additives. The invention likewise relates to concentrates and mineral oil middle distillates
30 containing these mixtures.

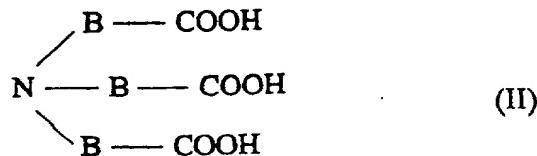
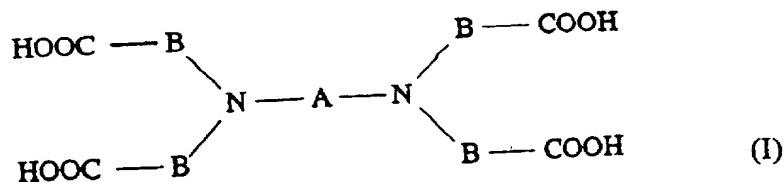
COMPONENT (a)

Component (a) is a reaction product of a poly(C_{2-20} -carboxylic acid) having at least one tertiary amino group with secondary amines.
35

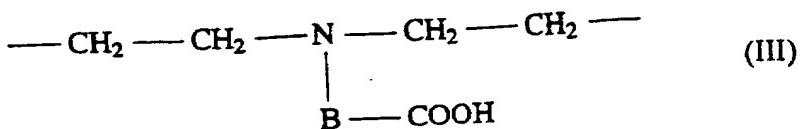
The polycarboxylic acid preferably contains at least 3, particularly preferably 3 to 12, in particular 3 to 5,

carboxyl groups. The carboxyl groups in the polycarboxylic acid preferably have 2 to 10 carbon atoms, acetyl groups being preferred. The carboxyl groups are linked in a suitable manner to the groups 5 polycarboxylic acid, for example via one or more C and/or N atoms. They are preferably bonded to tertiary nitrogen atoms which, in the case of a plurality of nitrogen atoms, are linked via hydrocarbon chains.

- 10 Component (a) is preferably an amide, amidoammonium salt or ammonium salt, or a mixture thereof, of aminoalkanecarboxylic acids of the formulae I and II



- 15 where A is straight-chain or branched alkylene of 2 to 6, preferably 2 to 4, in particular 2 or 3, carbon atoms, or a radical of the formula (III)



- 20 where B is a radical of 1 to 19 carbon atoms, preferably a C_{1-19} -alkylene radical, particularly preferably C_{1-10} -alkylene, in particular a methylene radical. A is preferably an ethylene radical.

- 25 The secondary amine may be selected from a multiplicity of amines which carry hydrocarbon radicals which may be bonded to one another.

The secondary amine is preferably of the formula HNR₂, where radicals R independently are straight-chain aliphatic radicals, in particular alkyl of 10 to 30, preferably 14 to 24, carbon atoms. They preferably have
5 no hetero atoms or double or triple bonds. Preferably, the radicals R are identical.

The secondary amines can be bonded by means of amide structures or in the form of their ammonium salts to
10 the polycarboxylic acid, also partly by means of amide structures and partly in the form of the ammonium salts. Preferably, little or no free acid groups are present.

15 Preferably, the amines are bonded completely in the form of the amide structures.

The amides or amidoammonium salts or ammonium salts of, for example, nitrilotriacetic acid, ethylenediamine-
20 tetraacetic acid or 1,2-propylenediaminetetraacetic acid, are obtained by reacting the acids with from 0.5 to 1.5, preferably from 0.8 to 1.2, mol of amine per carboxyl group.

25 The reaction temperatures are from about 80 to 200°C, the resulting water of reaction being continuously removed for the preparation of the amides. However, the reaction need not be continued completely to the amide; rather, from 0 to 100 mol% of the amine used may be
30 present in the form of the ammonium salt. Particulary preferred amines are dioleylamine, dipalmitylamine, dicoconut fatty amine and dibehenylamine, in particular di-tallow fatty amine.

35 The novel components (a) of the mixture and their preparation are described in EP-A-0 398 101. The reaction product of one mole of ethylenediaminetetraacetic acid and four moles of hydrogenated di-tallow fatty amine is particularly preferred.

If required, small amounts of conductivity improvers in the form of salts, in particular of hydrocarbon-soluble carboxylic acids and sulfonic acids or their metal and 5 ammonium salts, may also be added to the components (a) of the mixture.

Component (b)

10 Preparation of the novel components (b) of the mixture is carried out in a manner known per se by reacting maleic anhydride with C₈₋₃₀-alkylamines, preferably primary C_{8-C₁₈}-alkylamines, in a molar ratio of 1:1 at from 70 to 100°C by the process described in
15 DE-A-11 49 843 and EP-A-0 106 234; suitable primary amines are all amines defined within these limits, for example straight-chain or branched octyl-, nonyl-, decyl-, undecyl-, dodecyl-, tridecyl-, tetradecyl-, pentadecyl-, hexadecyl-, heptadecyl- and octadecylamine
20 and mixtures of these amines. The reaction product of one mole of maleic anhydride and one mole of tridecylamine is particularly preferred.

The novel mixtures can be prepared by simple mixing of
25 the components (a) and (b); these mixtures are added to the mineral oil distillates in amounts of 10-1000 ppm, preferably from 50 to 500 ppm. The amount of the component (a) is from 5 to 95, preferably from 30 to 95, in particular from 50 to 90, % by weight and the
30 amount of the component (b) is from 5 to 95, preferably from 5 to 70, in particular from 10 to 50, % by weight.

The novel polymer mixtures are used as additives for
35 mineral oil middle distillates, which are understood as meaning petroleum, light fuel oils and diesel fuels having a boiling point of from about 150 to 400°C. The polymer mixtures may be added to the middle distillates directly but are preferably added as a 20 to 70% by weight strength solution (concentrate). Suitable

solvents are aliphatic or aromatic solvents, such as xylene or mixtures thereof, and furthermore high-boiling aromatics mixtures, such as solvent naphtha, and middle distillates. The amount of mixture in the
5 mineral oil middle distillates is as a rule from 10 to 10,000, preferably from 20 to 5000, particularly preferably from 50 to 1000, ppm.

As a rule, the middle distillates also contain flow
10 improvers, for example based on ethylene/vinyl carboxylate copolymers. Depending on the intended use, the middle distillates may additionally contain further additives, e.g. conductivity improvers, corrosion-inhibiting additives, lubricity additives, anti-
15 oxidants, metal deactivators, antifoams, demulsifiers, detergents, cetane number improvers and/or dyes and fragrances.

The novel mixtures result in a substantial improvement
20 in the low-temperature flow properties in middle distillates, regardless of their origin, in that they effectively keep the paraffin crystals which separate out in suspension so that there is no blocking of filters and pipes by paraffin which has settled out.
25 They have a broad action and thus ensure that the paraffin crystals which have separated out are very well dispersed in various middle distillates; at the same time, they help to improve the lubricity of the middle distillates.

30 The effect of the combination of the components (a) and (b) is substantially better than the effect of the individual components at the same dose.

35 The examples which follow illustrate the invention.

EXAMPLES:

Example 1 (component (a))

240 g (0.48 mol) of hydrogenated di-tallow fatty amine (Armeen® 2HT from Akzo) and 35 g (0.12 mol) of ethylenediaminetetraacetic acid were melted and were
5 heated to 190°C, the resulting water of reaction being distilled off continuously. The reaction was terminated after about 25 hours at an acid number of less than 10 and an amine number of less than 1.1. By applying reduced pressure from a waterjet pump (2 hours, 120°C),
10 the water of reaction was removed completely. 265 g of a light brown, waxy solid were obtained. The product obtained was diluted with solvent naphtha so that the solids content of the product was 50% by weight.

15 Example 2 (component (b))

A mixture of 98 g (1.0 mol) of maleic anhydride and 199 g (1.0 mol) of tridecylamine was heated in 250 ml of solvent naphtha at about 70°C for 2 hours while
20 stirring. The light brown, clear solution obtained was then diluted with solvent naphtha so that the solids content of the product was 50% by weight.

Use Examples:

25 The novel mixtures were tested in two commercial German winter diesel fuels which complied with European diesel fuel standard EN 590; they are designated as D1 and D2 and are characterized by the following physical data:
30

	D 1 (97/526)	D 2 (96/86)
cloud point (°C) according to ISO 3015:	-6	-6
CFPP (°C) according to EN 116:	-10	-8
Density at 15°C (kg/m³) according to ASTM D 4052:	841	834
Sulfur content (ppm) according to EN 24260:	160	200
WS1.4 (µm) according to CEC F-06-A-96:	542	
Distillation according to ISO 3405:		
Initial boiling point (°C)	170	171
5% boiling point (°C)	203	193
10% boiling point (°C)	215	200
20% boiling point (°C)	235	212
50% boiling point (°C)	280	249
70% boiling point (°C)	308	282
90% boiling point (°C)	347	329
95% boiling point (°C)	364	345
Final boiling point (°C)	371	360

The following mineral oil middle distillate compositions were tested:

5

Mineral oil middle distillate compositions containing

1. as paraffin dispersant

- 10 one of the novel mixtures PD 1 (consisting of 83% by weight of component (a) from Example 1 and 17% by weight of component (b) from Example 2), PD 2 (consisting of 50% by weight of component (a) from Example 1 and 50% by weight of component (b) from Example 2), PD 3 (consisting of 67% by weight of component (a) from Example 1 and 33% by weight of
- 15

component (b) from Example 2) or PD 4 (consisting of 90% by weight of component (a) from Example 1 and 10% by weight of component (b) from Example 2) and, as comparative examples, the respective individual components from Example 1 and Example 2 or none of the components (Comparative Examples V2, V3, V1). V1 thus contains only MDFI as additive

10 and

2. as flow improver MDFI

15 a product based on ethylene/vinyl carboxylate which is marketed under the tradename Keroflux® ES 6100 by BASF AG.

Description of the test method:

20 The middle distillates were mixed with the amounts, stated in the table, of the novel mixtures PD 1 to PD 4 or of Examples 1 and 2 and of the flow improver MDFI at 40°C while stirring and then cooled to room temperature.

25 For the additive-containing middle distillate samples, the cold filter plugging point (CFPP) according to EN 116 was determined, and in some cases also the WS1.4 according to CEC-F-06-A-96.

30 The additive-containing middle distillates were cooled in 500 ml glass cylinders in a refrigerant bath from room temperature to -13°C at a cooling rate of about 14°C per hour and were stored at this temperature for 35 20 hours. The amount and appearance of the paraffin phase were then determined and assessed visually.

The cold filter plugging point (CFPP) according to EN 116 and the cloud point (CP) according to ISO 3015

were determined for the 20% by volume bottom phase separated off at -13°C from each sample. The smaller the deviation of the cloud point of the 20% by volume bottom phase from the original CP of the respective 5 middle distillate, the better are the paraffins dispersed.

The results obtained are shown in Tables 1 and 2:

10 Table 1: Dispersing experiments in D 1, CP: -6°C,
CFPP: -10°C, WS1.4: 542 µm

Mix-ture	Dose (ppm)	MDFI (ppm)	WS1.4 (µm)	CFPP (°C)	Paraf-fin sediment (% by vol- ume)	Dis- persed paraf-fins (% by vol ume)	20% bottom phase	
							CFPP (°C)	CP (°C)
PD 1	150	200		-30	10	90	-29	-6
PD 2	150	200	310	-25	10	90	-27	-6
PD 3	150	200		-26	10	90	-28	-6
PD 4	150	200	328	-28	10	90	-27	-5
V1	---	200	526	-25	44	0	-18	0
V2	150	200	394	-25	46	54	-20	-2
V3	150	200	369	-23	42	58	-23	-3

15 Table 2: Dispersing experiments in D 2, CP: -6°C,
CFPP: -8°C

Mix-ture	Dose (ppm)	MDFI (ppm)	CFPP (°C)	Paraffin sediment (% by volume)	Dispersed paraffins (% by volume)	20% bottom phase	
						CFPP (°C)	CP (°C)
PD 1	300	300	-23	6	94	-18	-5
PD 3	300	300	-20	8	92	-27	-5
PD 4	300	300	-29	8	92	-20	-5
V1	---	300	-22	30	0	-1	+3
V2	300	300	-30	10	90	-20	-3
V3	300	300	-20	30	70	-7	+2

The test results show that, in the mineral oil middle distillates, the novel mixtures result in a lower cloud point of the 20% bottom phase than the mixtures of the comparative examples.

5

This shows that, in mineral oil middle distillates, regardless of their origin, the precipitated paraffin crystals are effectively kept in suspension by the novel mixtures of the components (a) and (b) so that 10 filters and pipes are not blocked by paraffin which settles out. The novel mixtures have a very broad action and ensure that the paraffin crystals which separate out are very well dispersed in various middle distillates.

15

At the same time, the tests results show that, at the same total dose, the novel mixtures lead to a lower WS1.4 and hence to improved lubricity of low-sulfur diesel fuels then the respective individual components.

CONFIDENTIAL

09/830149

ପ୍ରକାଶ ମନ୍ତ୍ରୀଙ୍କ ପରିଷଦ୍ୟ

23 APR 2001

- 14 -

O.Z. 0050/49464

Claims as enclosed to IPER

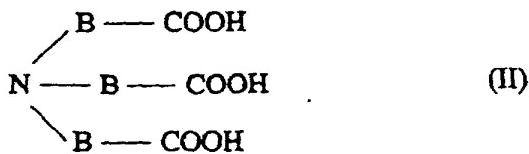
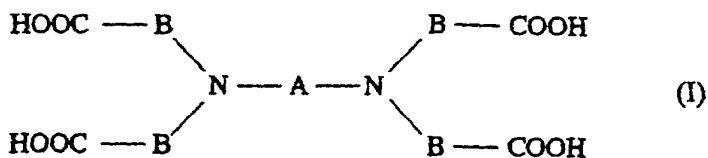
5 We claim:

- ### 1. A mixture containing

10 (a) from 5 to 95% by weight of at least one reaction product of a poly(C₂₋₂₀-carboxylic acid) having at least one tertiary amino group with secondary amines and

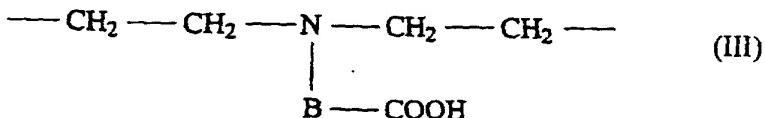
15 (b) 5-95% by weight of at least one reaction product of maleic anhydride and a primary alkylamine.

2. A mixture as claimed in claim 1, wherein the
poly(C₂₋₂₀-carboxylic acid) having at least one
tertiary amino group is of the following formula I
or II



25

where A is a straight-chain or branched C₂₋₆-alkylene radical or a radical of the formula III



where B is a C₁₋₁₉-alkylene radical.

3. A mixture as claimed in claim 1 or 2, wherein the reaction product (a) is an amide, amidoammonium salt or ammonium salt in which no carboxyl group, one carboxyl group or a plurality of carboxyl groups is or are converted into amido groups.
- 5
4. A mixture as claimed in any of claims 1 to 3, wherein the secondary amine of the component (a) is of the formula HNR₂, where R is straight-chain or branched C₁₀₋₃₀-alkyl.
- 10
5. A mixture as claimed in any of claims 1 to 4, wherein the primary alkylamine of the component (b) is a C₈₋₃₀-alkylamine.
- 15
6. The use of a mixture as claimed in any of claims 1 to 5 as an additive for mineral oil middle distillates.
- 20
7. The use as claimed in claim 6 as a paraffin dispersant and lubricity additive for mineral oil middle distillates.
- 25
8. A mineral oil middle distillate containing a mixture as claimed in any of claims 1 to 5.
9. A mineral oil middle distillate as claimed in claim 8, furthermore containing flow improvers, conductivity improvers, corrosion-inhibiting additives, lubricity additives, antioxidants, metal deactivators, antifoams, demulsifiers, detergents, cetane number improvers, dyes, fragrances or mixtures thereof.
- 30
10. A concentrate containing 10-60% by weight, based on the total amount of the concentrate, of a
- 35

mixture as claimed in any of claims 1 to 5,
dissolved in the hydrocarbon solvent.

11. A mineral oil middle distillate as claimed in
5 claim 8, further containing flow improvers based
on ethylene/vinyl carboxylate copolymers.

Abstract

The mixture contains

5

(a) from 5 to 95% by weight of at least one reaction product of a poly(C₂₋₂₀-carboxylic acid) having at least one tertiary amino group with secondary amines and

10

(b) from 5 to 95% by weight of at least one reaction product of maleic anhydride and a primary alkylamine.

15

CONFIDENTIAL COPY

Declaration, Power of Attorney

Page 1 of 3

0050/049464

We (I), the undersigned inventor(s), hereby declare(s) that:

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

Paraffin dispersants having a lubricity effect for mineral oil middle distillates

the specification of which

[] is attached hereto.

[] was filed on _____ as

Application Serial No. _____

and amended on _____.

[x] was filed as PCT international application

Number _____

on 19/10/1999

and was amended under PCT Article 19

on _____ (if applicable).

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application(s)

Application No.	Country	Day/Month/Year	Priority Claimed
19848621.9	Germany	21 October 1998	[x] Yes [] No

We (I) hereby claim the benefit under Title 35, United States Codes, § 119(e) of any United States provisional application(s) listed below.

(Application Number) _____ (Filing Date) _____

(Application Number) _____ (Filing Date) _____

We (I) hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

Application Serial No.	Filing Date	Status (pending, patented, abandoned)
_____	_____	_____
_____	_____	_____
_____	_____	_____

And we (I) hereby appoint: Norman F. Oblon, Reg. No. 24,618; Marvin J. Spivak, Reg. No. 24,913; C. Irvin McClelland, Reg. No. 21,124; Gregory J. Maier, Reg. No. 25,599; Arthur I. Neustadt, Reg. No. 24,854; Richard D. Kelly, Reg. No. 27,757; James D. Hamilton, Reg. No. 28,421; Eckhard H. Kuesters, Reg. No. 28,870; Robert T. Pous, Reg. No. 29,099; Charles L. Gholz, Reg. No. 26,395; William E. Beaumont, Reg. No. 30,996; Jean-Paul Lavallee, Reg. No. 31,451; Stephen G. Baxter, Reg. No. 32,884; Richard L. Treanor, Reg. No. 36,379; Steven P. Weihrouch, Reg. No. 32,829; John T. Goolkasian, Reg. No. 26,142; Richard L. Chinn, Reg. No. 34,305; Steven E. Lipman, Reg. No. 30,011; Carl E. Schlier, Reg. No. 34,426; James J. Kulbaski, Reg. No. 34,648; Richard A. Neifeld, Reg. No. 35,299; J. Derek Mason, Reg. No. 35,270; Surinder Sachar, Reg. No. 34,423; Christina M. Gadiano, Reg. No. 37,628; Jeffrey B. McIntyre, Reg. No. 36,867; William T. Enos, Reg. No. 33,128; Michael E. McCabe, Jr., Reg. No. 37,182; Bradley D. Lytle, Reg. No. 40,073; and Michael R. Casey, Reg. No. 40,294; our (my) attorneys, with full powers of substitution and revocation, to prosecute this application and to transact all business in the Patent Office connected therewith; and we (I) hereby request that all correspondence regarding this application be sent to the firm of **OBLON, SPIVAK, McCLELLAND, MAIER & NEUSTADT, P. C.**, whose Post Office Address is: Fourth Floor, 1755 Jefferson Davis Highway, Arlington, Virginia 22202.

Customer No. 21425

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

1 - 00 Bernd Wenderoth
NAME OF INVENTOR

Bernd Wenderoth

Signature of Inventor

Date 5/03/2001

Römerstr.115c
68623 Lampertheim DEX
Germany
Citizen of: Germany

Post Office Address: same as residence

2 - 00 Dieter Hermeling
NAME OF INVENTOR

Dieter Hermeling

Signature of Inventor

Date 5/03/2001

Am Wasserturm 19
67459 Böhl-Iggelheim DEX
Germany
Citizen of: Germany

Post Office Address: same as residence